EFCOG 2019 – NSR&D Subgroup Meeting

NSRD-12/20 – Novel Mini-Tubular HEPA Media for Nuclear Facility Ventilation Systems

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Goals: Lower Costs for Nuclear Facilities, Maintain Safety

Destroyed filter bank after a fire

Water damage to filters following a fire

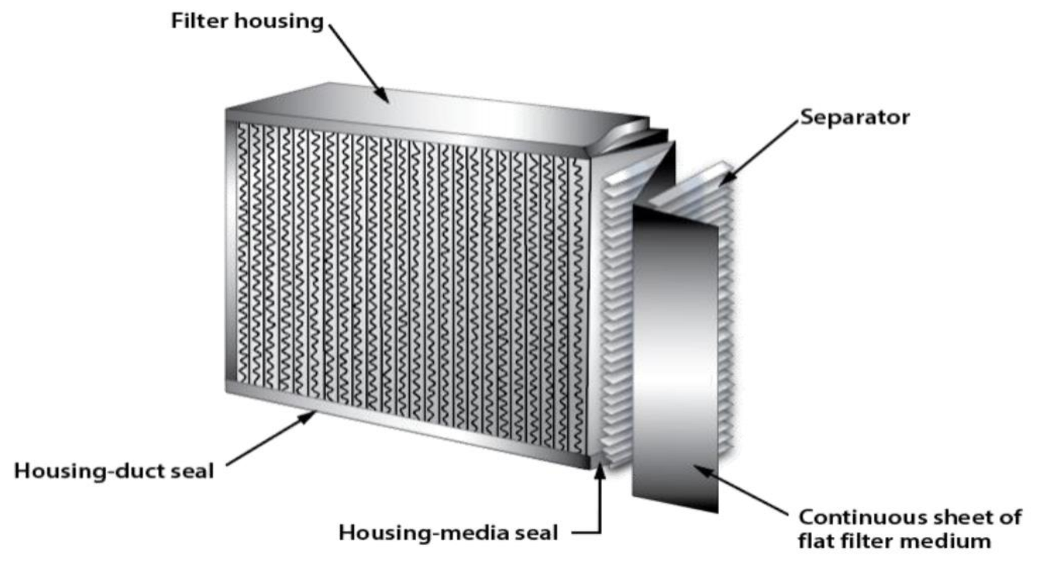
Ceramic filters perform at higher temperatures and are likely to eliminate reliance on credited fire suppression systems

Conventional Filters

DOE Needs Analysis:

- 100% of knowledgeable nuclear air cleaning professionals believe HEPA filter media strength is very, or extremely, important
- 92% believe it is important to develop alternatives to current glass-fiber filters

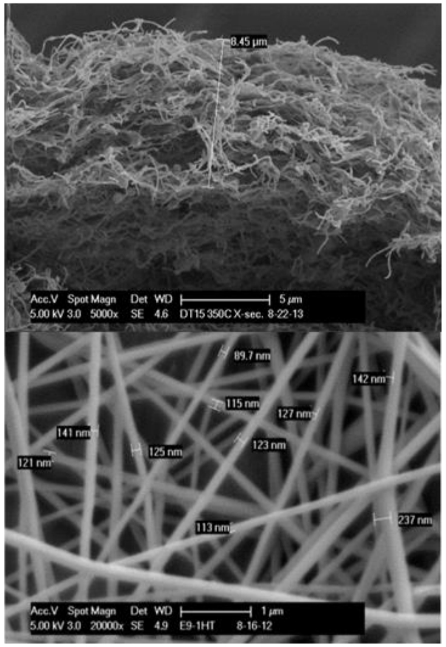
Components of a HEPA filter



Susceptible to temperature, water, bursting

High-Temperature Filter Media

Thermal condition	Standard Fiberglass Filters	Mini-Tubular Filter Media
Room temperature	Moisture degrades fiberglass	Potentially moisture resistant
Intermediate temperature	Lose organic binders	No organic binders
High temperature	Softening and crystallization of fiberglass	Stable



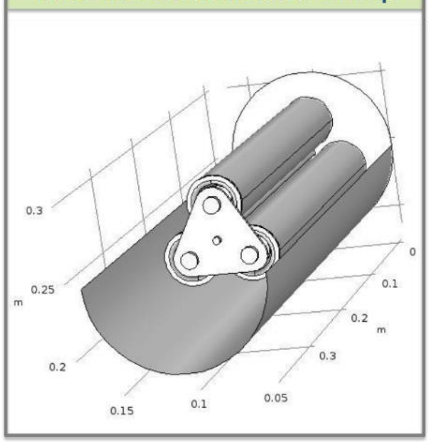
- Nanofiber mini-tubular filter media are promising
  - Higher filtration efficiency, lower pressure drop with decreasing fiber diameter
  - Use of mini-tubular approach is novel, with new challenges, but can be used to avoid existing challenges due to substrate constraints and wide-area deposition of ceramic nanofibers

Current R&D focused on overcoming shrinkage problems and reducing pressure drop

LLNL Ceramic Filter Development

Filter prototype of patented design

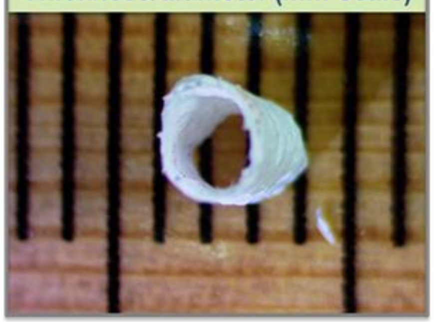
Filter tubes with media overwrap



- Ceramic Filters
  - Ceramic HEPA Filters
    - Tube Overwrap
      - Conventional Media
      - Nanofiber Media
    - MTC Media
    - Sealants
    - Advanced Manufacturing and Sensing
  - Ceramic Prefilters

Tube length (mm-scale)

Thickness/diameter (mm-scale)



MTC Media Development

Purpose: To improve the safety of DOE nuclear facilities in fire scenarios, create advanced ceramic filtration components that can survive a fire event while reducing pressure drop, increasing performance, and reducing costs

Benefit: Our engineering solution protects filters during a fire to simplify and reduce the cost of safety- and filter-support systems for operations

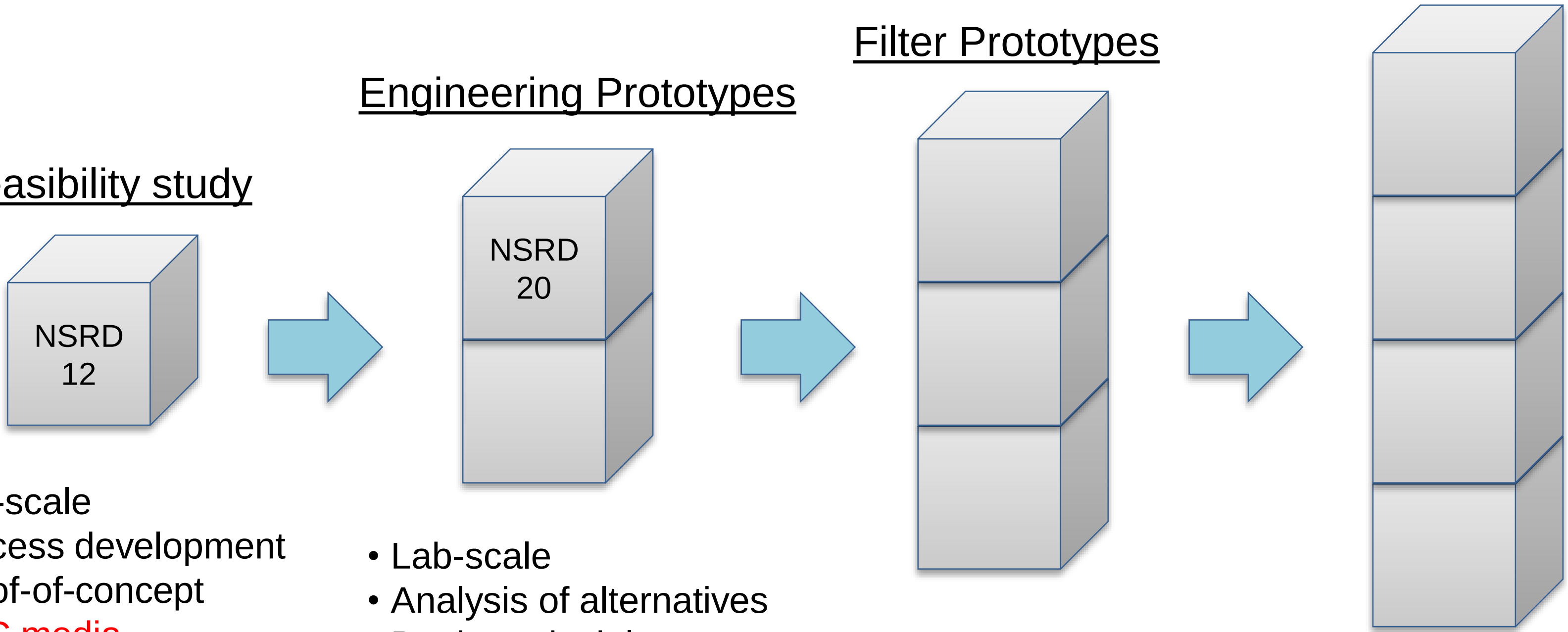
NSRD-12

- Demonstrate MTC media can reduce pressure drop
- Develop processes to produce MTC nanofiber filtration media

NSRD-20

- Prepare MTC media using different manufacturing approaches
- Test pressure drop and filtration efficiency of MTC filter elements

MTC HEPA Filter Media Development



Feasibility study

- Lab-scale
- Process development
- Proof-of-concept
- MTC media

Engineering Prototypes

- Lab-scale
- Analysis of alternatives
- Design principles
- Process optimization
- Tooling development
- MTC filter element

Filter Prototypes

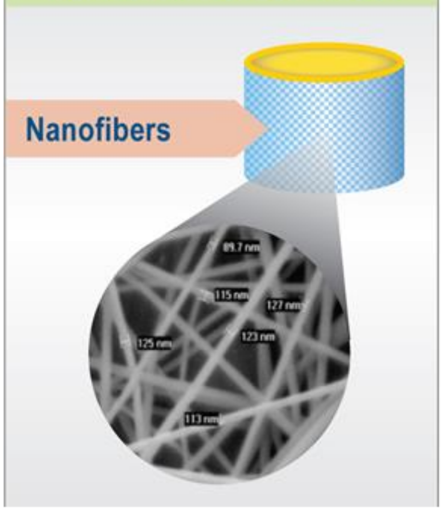
- Scaling
- Automation
- Integration
- Certification
- MTC filter

Manufacturing

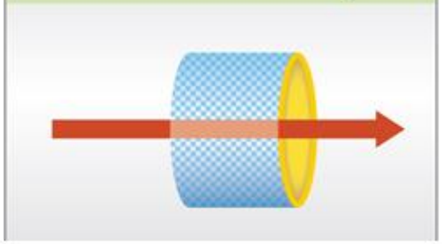
- Licensing
- Deployment
- Servicing
- Continuous improvement

MTC HEPA Media – Reduced dP Proof-of-Concept

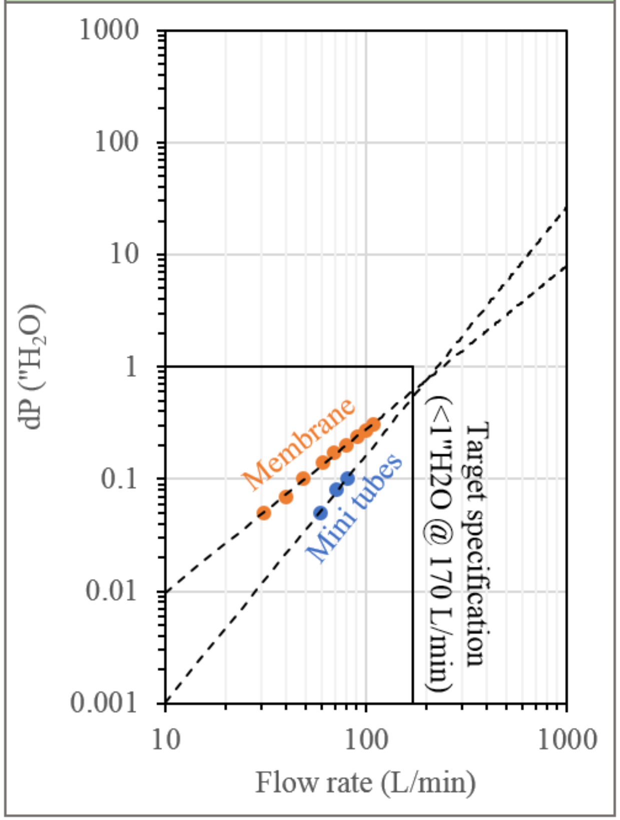
Individual Mini-Tubular Media



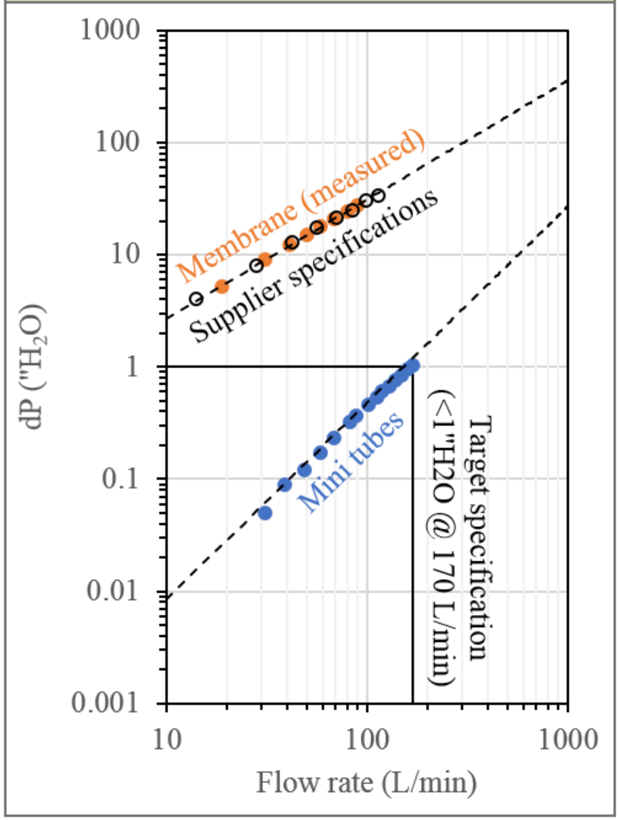
Contact filtration flow path



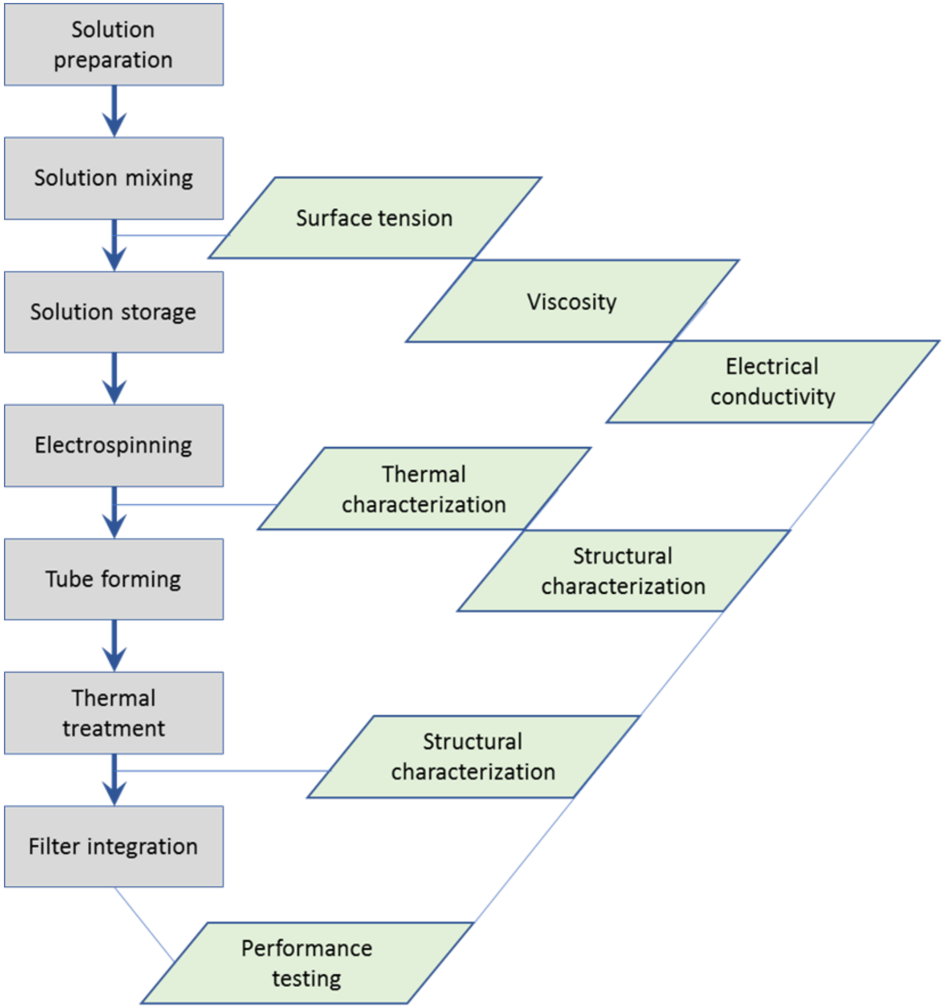
Support media surrogate filter performance



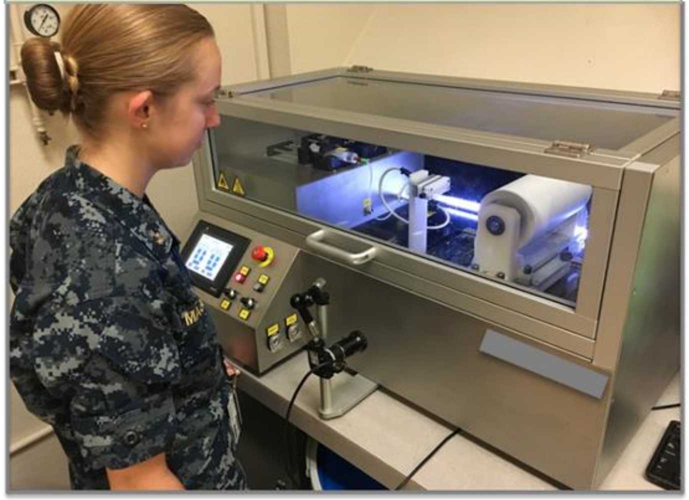
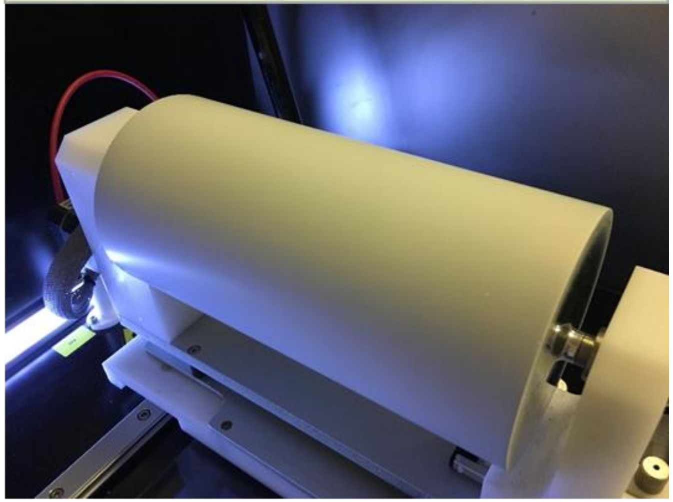
HEPA media surrogate filter performance



MTC HEPA Filter Media – Process Development

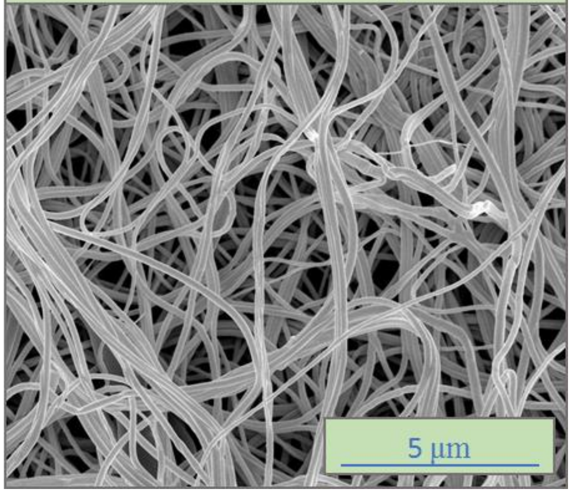


Electrospinning Process

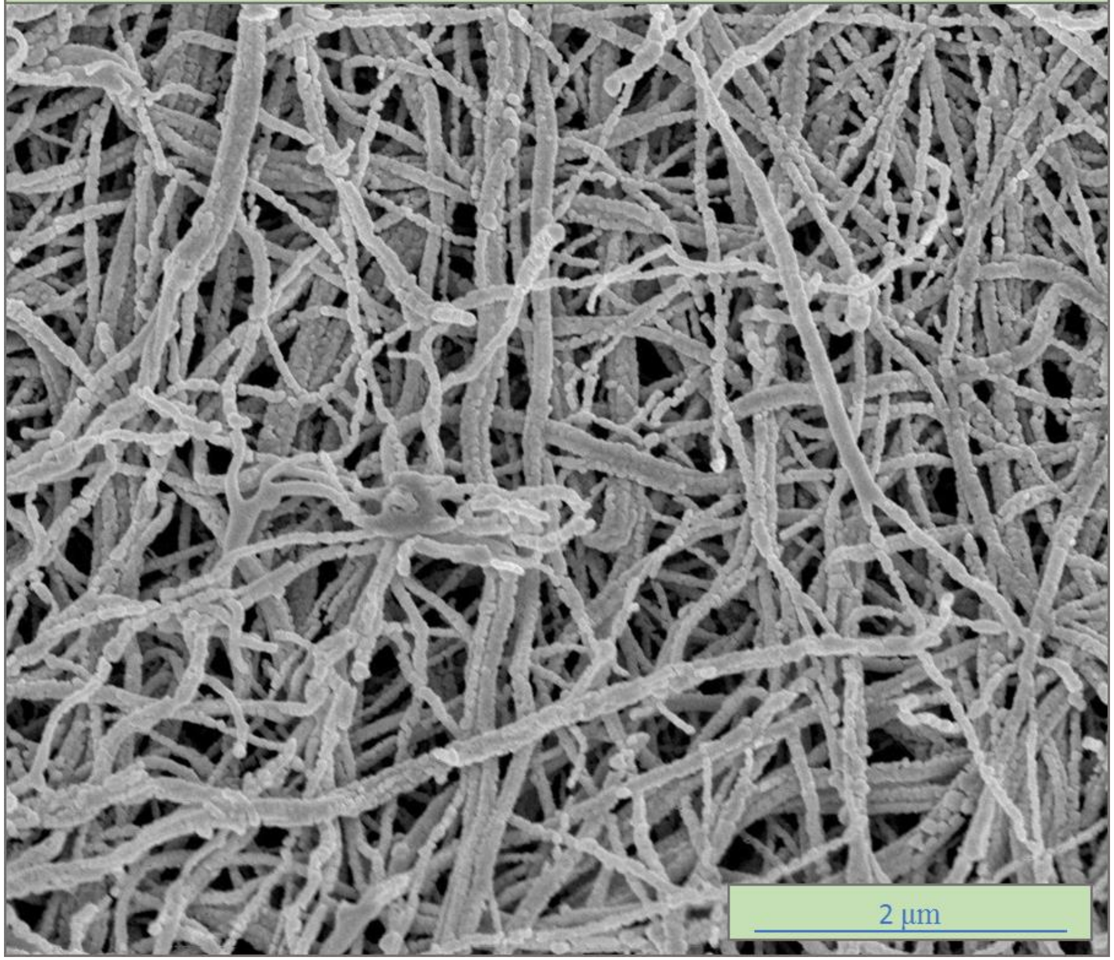
Electrospinning equipment	Fiber jet	Fiber collector
		

Pre-ceramic and Ceramic Electrospun Nanofibers

Pre-ceramic nanofibers (System 2)



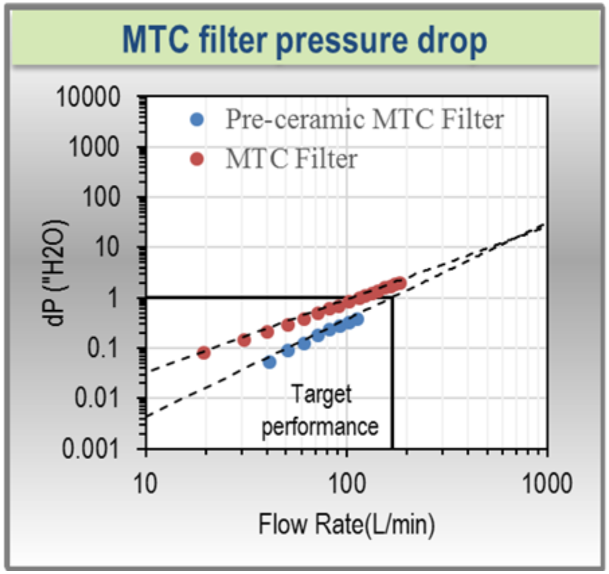
Ceramic nanofibers produced from System 2 and after 1000°C heat treatment

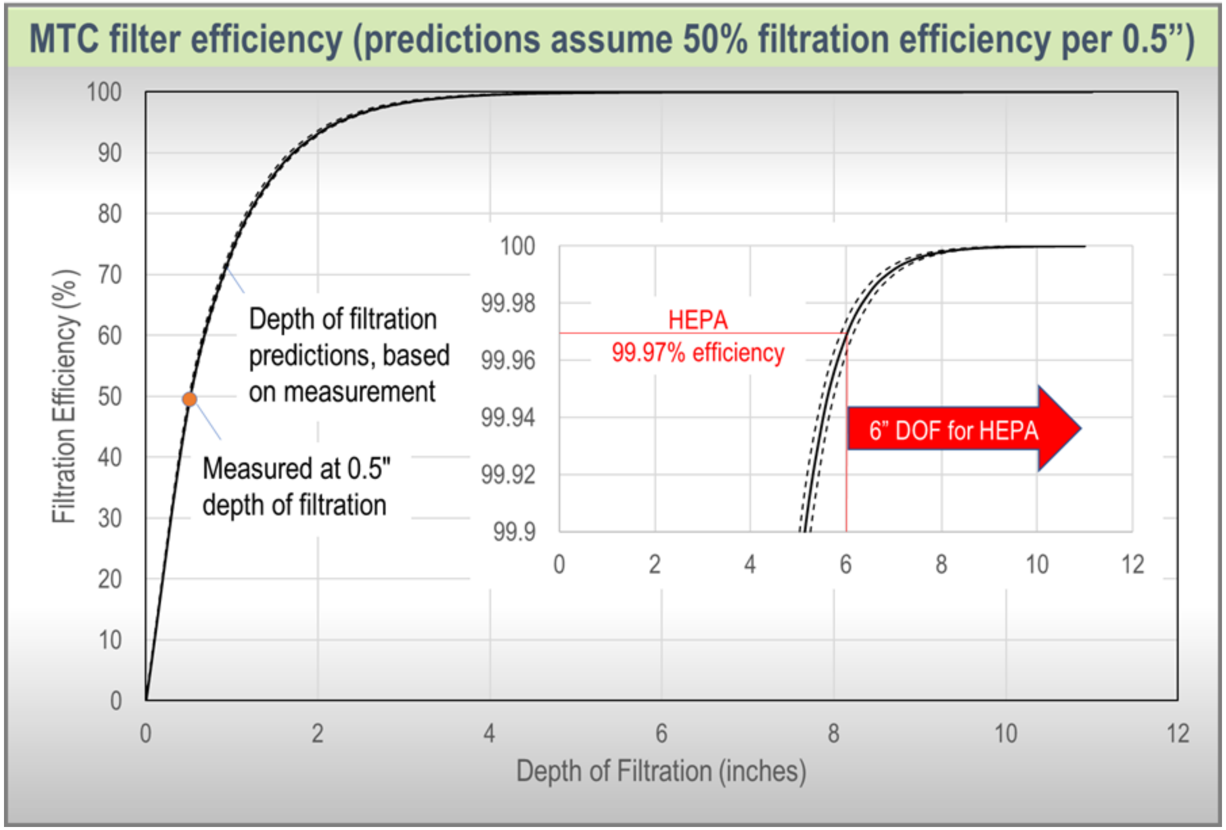


140 nm avg

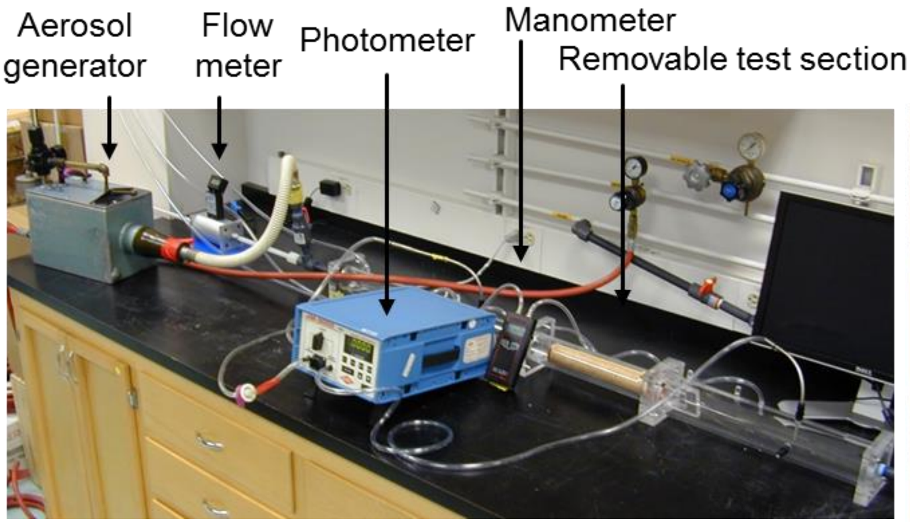
80 nm avg

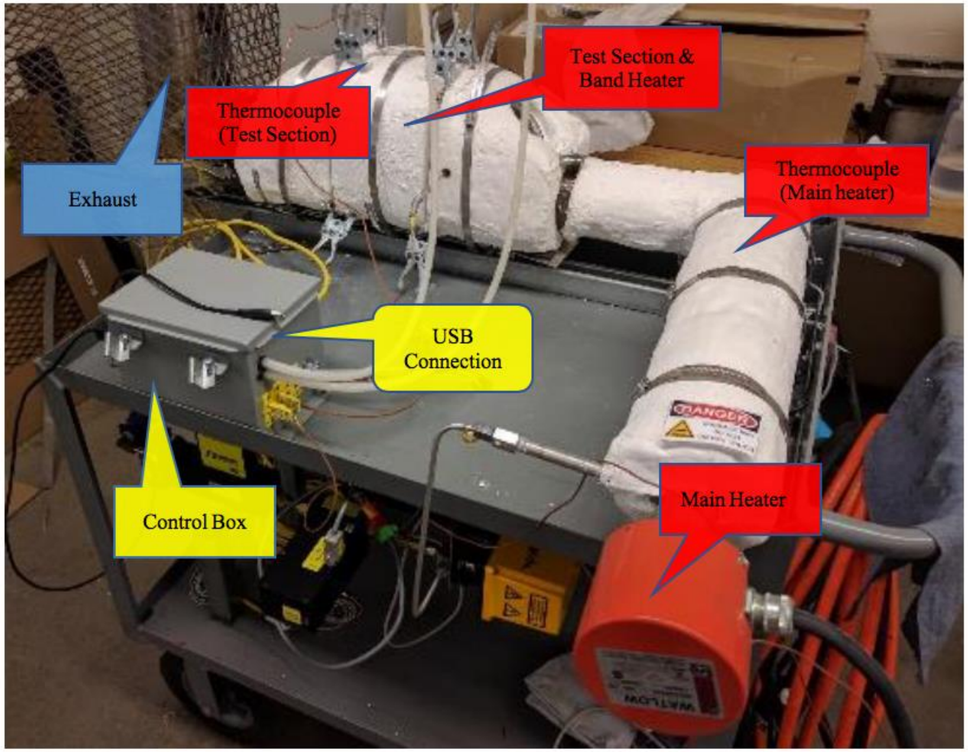
MTC HEPA Media Proof-of-Concept



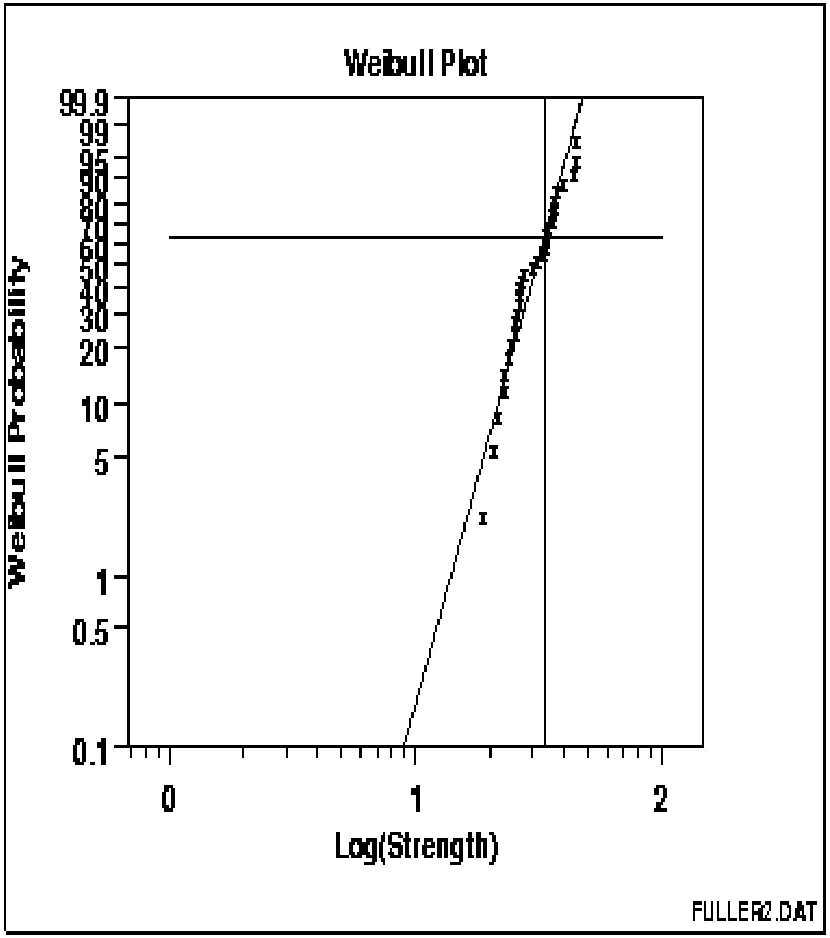


Filter Testing Capabilities at LLNL

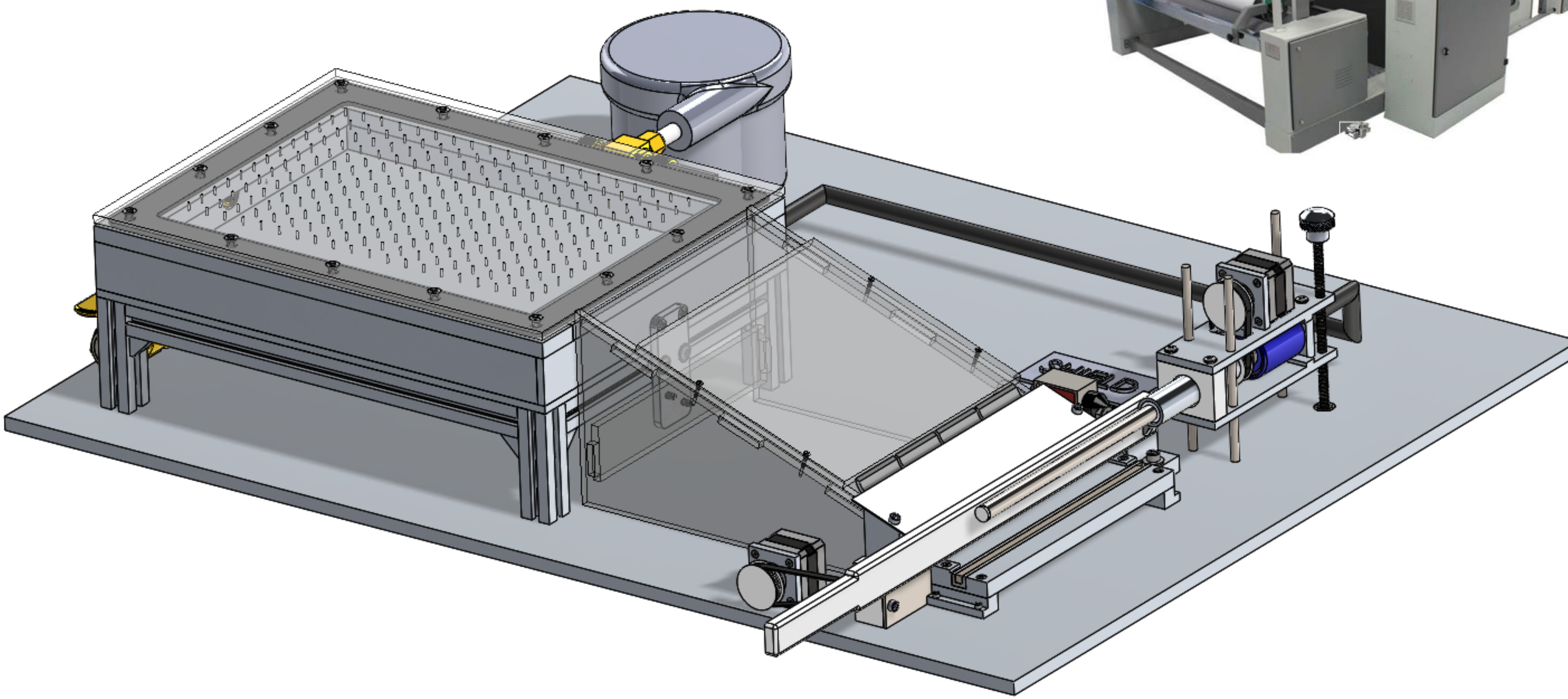




Process Optimization

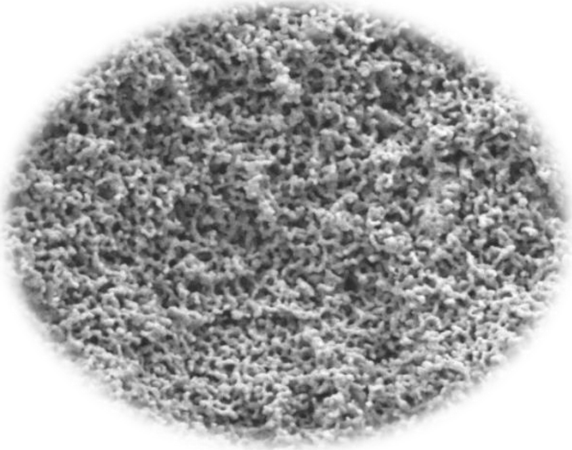


Automating MTC Production - Tooling Development by Cal Poly



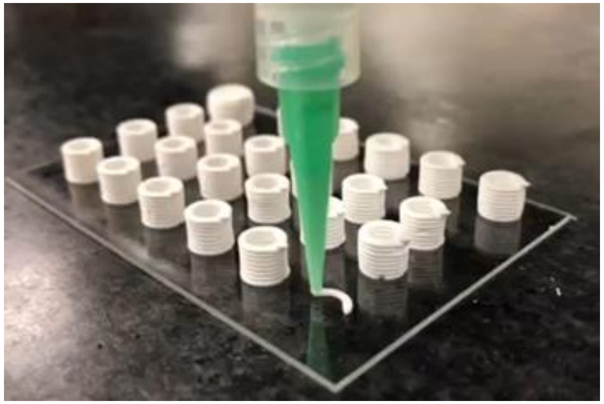
Analysis of Alternatives: Current & Future

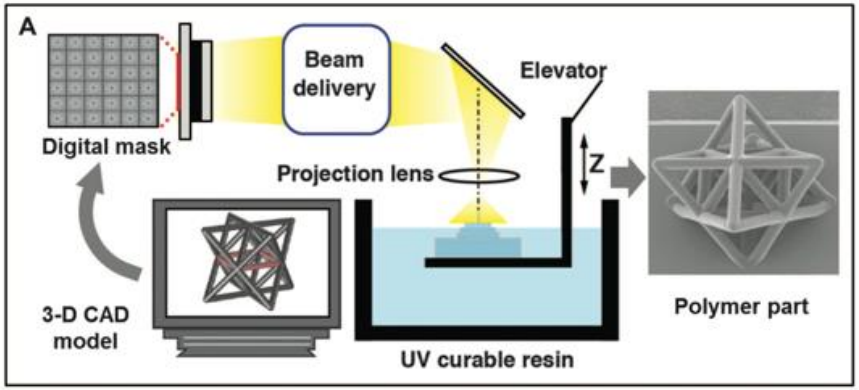
Extruded tubes:



Future Work may include
Projection Micro Stereo Lithography
(PμSL)

DIW:





Acknowledgements

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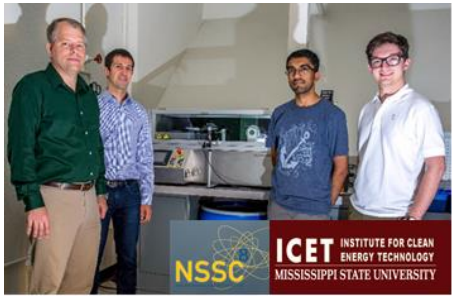

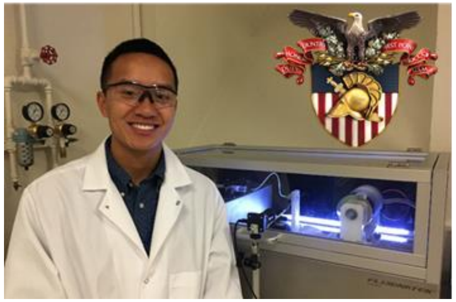

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  - Brandon Bogle
- MARA (Barry Goldman)
  - Jack Bui, West Point
  - Jamie Maguire, Naval Academy
- NSSC (Prof. Chintalapalle)
  - Nanthakishore Makeswaran

Background and Q&A

Design and Development of Electro-Spun Nanoarchitectures of Materials for Extreme Environment Applications
UTEP-LLNL Collaborative Program

Nanthakishore Makeswaran, University of Texas-El Paso, Department of Engineering, 500 W. University Blvd., El Paso, TX 79968
Email: nmakeswaran@miners.utep.edu

Goals and Objectives


Ultrasensitive chemresistor

The goal of this project is to develop electro-spun Ga_2O_3 nanofibers and determine their effectiveness as particulate air filters and oxygen sensors in extreme temperature environments. Upon completion of the project the expected deliverable will be a Ga_2O_3-based oxygen sensor that have the potential to operate in extreme environments.

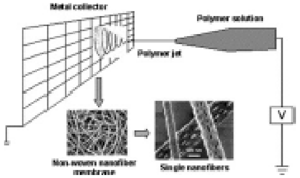
Introduction

Electrospinning is an efficient and scalable manufacturing technology for the production of a wide variety of nanofiber materials, including several materials for sensor applications.


Industrial scale electrospinning equipment

This research focuses around nanoscale fibers (50-500 nm) derived from a polymer solution with ceramic precursors. These structures offer two key advantages. They provide superior mechanical performance, specifically stiffness and tensile strength. They also possess very high surface to volume ratios that are expected to improve sensitivity and reaction time in sensor applications.

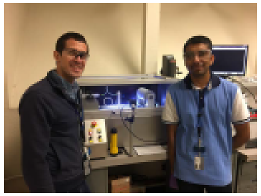
The jets of the solution form what is known as a Taylor Cone when a critical voltage is reached, overcoming the solution's viscosity. The fibers are extracted, elongated, and deposited on the collector.

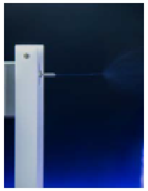


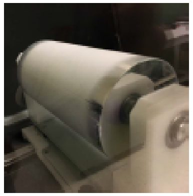
Overview of electrospinning process

Methods

To produce Ga_2O_3 nanofibers, a Ga precursor is introduced into the polymer solution for spinning. The fibers are collected and then calcined at high temperatures (typically in excess of 700°C), burning away the polymer and leaving behind ceramic nanofibers. A polymer solution of water, ethanol, polyvinylpyrrolidone (PVP), $Ga(NO_3)_3$ were chosen for its ease of preparation and minimal hazards during handling. Previous research at LLNL highlighted the importance of selecting the formulation and careful preparation procedures. The polymer/gallium precursor solution is then spun in a lab-scale commercial electrospinning system and deposited on a rotating drum collector. Experiments on varying parameters in a custom laboratory-scale system were conducted before scaling up to the commercial system.

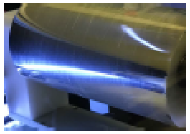

Electrospinning equipment


Fiber jet

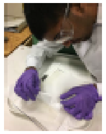

Nanofiber collector

Major Findings

Initial electrospinning of pure polymer solution produced a uniform fiber mat. Introduction of Ga precursors, however, led to few, sporadically developing fibers and minimal deposition. Adjustments led to deposition, but with sporadic spraying and brittle fiber mats. Further adjustments enabled successful deposition and removal.

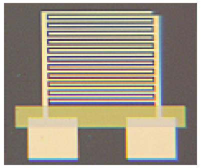

Lack of fiber deposition


Deposition, but with spraying


Successful removal!

Next Steps

The calcining process to convert the precursors to Ga_2O_3 fibers needs to be developed. Successfully calcining the nanofibers will enable integrating them into sensor devices and filtering media to determine oxygen sensing performance and to determine how well they filter airborne particulates and to determine how well they filter airborne particulates. The sensitivity and response time of the sensor will be tested.


Interdigitated electrode configuration for sensor device (Case Western University)


3D printing approach to position nanofiber mats in a filter

Conclusion

The project to develop and test nano-scale fibers for use in extreme environments is still ongoing. Initial experimentation led to results inconsistent with established literature, but modification to the materials have been made and new experiments are on track to develop usable nanofibers. The research producing these fibers, with their uses as gas sensors and/or filters, will help improve safe operations at nuclear facilities as well as provide technology for other defense and commercial applications. Future work includes examining additional potential detector applications of interest to the nuclear industry and other extreme environments.

Acknowledgements

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Unclassified

Transient Heating and Thermomechanical Stress Modeling of Ceramic HEPA Filters under Fire Conditions

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Lawrence Livermore National Laboratory

U.S. DEPARTMENT OF ENERGY | Office of Science

MISSISSIPPI STATE UNIVERSITY

Abstract

Next generation HEPA filter technology developed at LLNL is designed to survive fire scenarios using ceramics and advanced materials. The transient heating and thermomechanical stresses of the filter during a fire were modeled using COMSOL and considering 500°C inlet air flowing at a rate of 6 CFM. The models progressed from simple 2D sub-sections to 3D assemblies

Introduction

- High-Efficiency Particulate Air (HEPA) Filters capture biological, chemical and radiological aerosols
- Fire and water damage has been a problem in the past for filters
- Traditional Fibrous filters are susceptible to structural and operational failure due to fire, water, corrosives, etc.

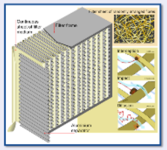

Figure 1: Filter cross-section and methods of particle collection (Air Purifier Rating)

Figure 2: Rocky Flats HEPA filter bank fire damage (Department of Energy)

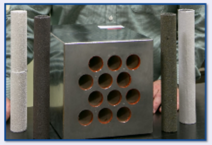

Figure 3: Ceramic HEPA filter housing assembly with individual filters (LLNL)

COMSOL workflow process

Create Geometries → Specify Material Properties → Define Physics Models and Initial Conditions → Create Mesh → Choose and Run Study → Post Process Results

Workflow

Geometries

- 2D simplified geometries utilized manual geometry input
- Complex 3D geometries employed COMSOL's CAD import module for SOLIDWORKS assemblies
- Symmetry enabled computation simplification

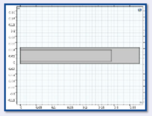

Figure 4: 2D asymmetric simplified model of filter

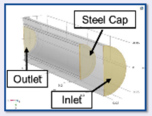

Figure 5: 3D simplified filter model with symmetry plane.

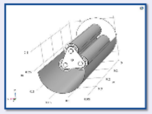

Figure 6: 3D imported CAD assembly model

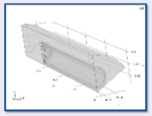

Figure 7: 3D imported CAD assembly model utilizing symmetry to reduce computation

Material Properties

- Integrated and prescribed scalar and variant COMSOL material library properties for each component
- Materials: **Air, 410 Stainless Steel, Porous Ceramic**
- Properties: *Density, Viscosity, Permeability, Porosity, Heat Capacity, Elastic Modulus, Poisson's ratio, Thermal Expansion Coefficient, Specific Heat, and Thermal Conductivity*

Workflow (continued)

Physics Models

- **Turbulent Flow**
  - Turbulent flow conditions experimentally determined
  - Accounts for flow through porous media
  - Compared computed flow to experimental measurements
- **Heat Transfer**
  - Couples with Turbulent Flow for convective and conductive heating
  - Accounts for heat transfer in porous media
  - Time-dependent heating ramp function
- **Structural Mechanics**
  - Couples structural response from Heat Transfer for thermomechanical stress

Mesh

- Mesh verification study on 2D and 3D models

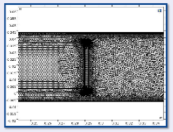

Figure 8: 2D simplified model with 'Finer' mesh applied

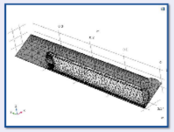

Figure 9: 3D symmetry model with 'Extra Coarse' mesh

Study

- **Stationary**
  - Solved time independent Thermomechanical stress at 500°C
- **Time-Dependent**
  - Calculates Transient Heating over a 60 second period with 1 second intervals
  - Complexity of CFD increased computation time significantly

Results

Thermomechanical Stress

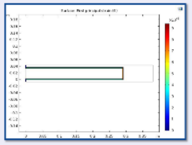

Figure 10: First Principal Strain plot of 2D model

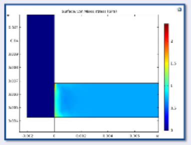

Figure 11: Stress and deformation plot at Steel Baseplate-Filter interface of 2D model

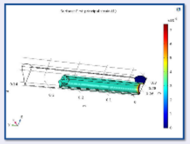

Figure 12: First Principal Strain plot of 3D model

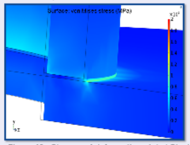

Figure 13: Stress and deformation plot at Steel Baseplate-Sealant-Filter interface of 3D model

- Stress magnitudes greatest at material interfaces
- Sealant experienced average stress less than 300 MPa
- Principle Strains similar in location but the magnitude is decreased for the 3D model

Results (continued)

Transient Heating

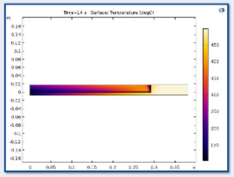

Figure 14: Heating Plot of 2D symmetry model at 14 s

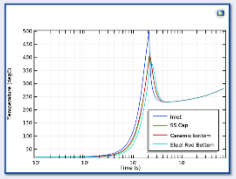

Figure 15: Average temperature plot for various locations of 3D model

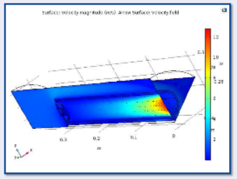

Figure 16: Velocity magnitude plot for 3D symmetry model

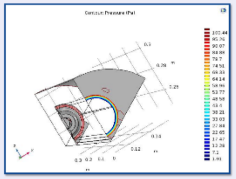

Figure 17: Pressure contour plot in porous mediafor 3D symmetry model

Experimental Data Comparison

- Compared 2D and 3D models differential pressure (dP) to experimental data
- Flow rates less than 6 CFM were laminar-to-turbulent flow transition zone

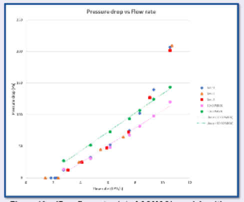

Figure 18: dP vs flow rate plot of COMSOL models with experimental flow data

Conclusion

- COMSOL provided thermal strain characterization for the ceramic tube-stainless steel interface for sealant design
- Estimated conservative heating rate for the filter assembly at 500°C fire conditions
- Filter heated to 90% inlet temperature in approximately 60 seconds
- Models agreed with relevant turbulent flow conditions at ≥ 6 CFM compared to experimental data

Future Work

- Consider incorporation of sealant design to evaluate thermal stress/strain reduction
- Utilize CFAST fire modeling to visualize filter heating conditions as function of position
- Continue to scale up models to full housing assemblies for heat loss to steel housing
- Use COMSOL to model additional design approaches

Design and Development of 3D Printed Ceramic Filter Media Architectures for Particulate Filtration

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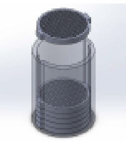
Goals and Objectives



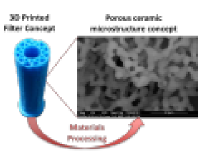
Example 3D printing approach to position nanofiber filter media mats in a filter

Several students explored potential opportunities for the use of 3D printing to develop new and innovative filter media architectures for particulate filtration. The goal is to explore several approaches for indirectly, and directly, 3D printing ceramic filter media. This promising technology produces complex shapes not possible by other methods and enables synergistic applications with other recent inventions and advances in materials engineering. The capabilities of several 3D printers, both commercial and at LLNL, are evaluated. This research highlights areas of opportunities and challenges.

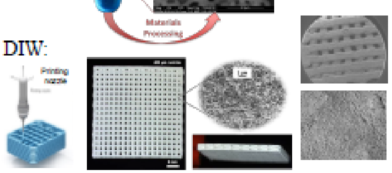
Introduction

Additive manufacturing is an emerging technology for potentially utilizing materials for particulate filtration. This can be done to indirectly, or directly, produce 3D printed porous ceramics. The indirect approach is to use commercial 3D printers, which often use polymeric materials, to print the architecture and then post-process the sample using a variety of techniques into a porous ceramic, e.g., via a ceramic coating. The direct approach is to use cutting-edge 3D printers, which can utilize additional materials, such as metals or ceramics. This can be done utilizing Direct Ink Write (DIW, middle) and projection microstereo-lithography (PμSL, bottom).

Indirect:

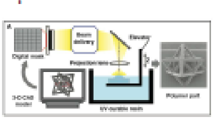


DIW:



- Self supported ink, shear thinning rheology
- Drying, curing and sintering of printed parts
- Multi-material capabilities

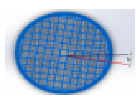
PμSL:



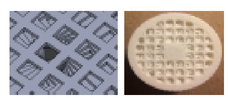
- UV-curable polymer resin
- Smaller feature size ~10 μm
- Layer thickness ~400 nm
- Lower viscosity inks
- More design freedom

3D Printing Lessons Learned

Students explored several approaches to the overall filter design (e.g., printing monoliths vs. printing modular layers of filter media, degree offset per layer, separation between layers) as well as the design for individual layers of filter media (e.g., grid patterns, shape of holes). Several commercial 3D printers at UC Berkeley, Cal Poly, and LLNL were utilized. A Type A Machine Series 1 3D printer at UC Berkeley Jacobs Hall was used primarily, as it is more efficient to utilize for testing basic designs than higher resolution printers. An analysis of higher resolution printers, as well as printers utilizing different technologies, is provided to benefit future projects. Early 3D printed filters explored the limits of commercial equipment (e.g., using ABS plastic, epoxy resin) and calculated pressure drop and porosity. Experiments determined that print designs can be optimized to maximize computational efficiency for manufacturing. Printer capabilities and limitations were identified by creating basic test prints that challenged the printer's ability to create complex geometries and print accurately. There are several approaches to overcoming the challenge of producing holes: 3D designs to produce holes, designing channels to facilitate ease of removal/flow guides, etching, removing filler material (e.g., dissolving, vibration). Several lessons learned on minimizing print failures are documented. A key finding is that the line width in a CAD drawing is widened significantly during 3D printing, thus true pore sizes are smaller than anticipated.



CAD depicting 10° offset between discs



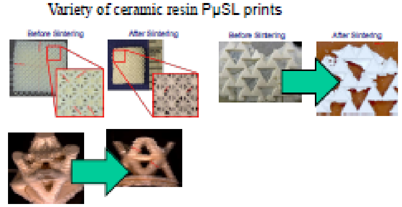
Close up of winding staircase: CAD (left), as 3D printed (right)

Example 3D prints: Cal Poly (top left), UC Berkeley (top right), LLNL (bottom)

Example print failure mechanism

Before & After Sintering

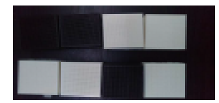
Variety of ceramic resin PμSL prints



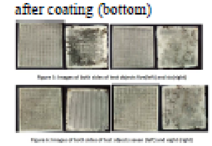
Extruded porous ceramic tubes before sintering

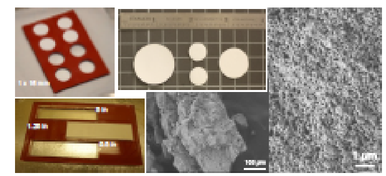
Indirect & Post Processing

Several post-processing are promising. Students focused on dip coating; other approaches have been identified for future research. Research on dip coating is facilitated by understanding potential issues with viscosity limits to coat a range of pore sizes. This is highlighted in research to develop a repeated protocol to uniformly coat internal complex geometries that can be produced by 3D printing, as well as to characterize the coatings themselves. A surrogate coating (latex patent) was utilized under a range of viscosities. Several test objects with line spacings on the order of 100 microns were designed and printed.

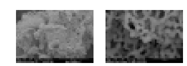


Print samples before (top) and after coating (bottom)





Sacrificial Polymer Template Method



Future research: vapor phases to control ceramic micro- and nano- porosity

Conclusion

Additive manufacturing enables complex filter designs, including designing macro- (3D print), and micro/nano-porosity (via processing). These advances pave the way for multi-functional filters (e.g., large particle removal, small-particle classification/collection, particle removal and classification, and/or concurrently filter particles and collect/treat gases). They can potentially be used where existing filters are used, such as HEPA filtration in ventilation systems, raw materials (e.g., particulate from oil), fluid feedstocks (e.g., slurries, molten metals), and waste streams. Additive manufacturing can be used to enable applications utilizing one device to replace duplicate systems utilizing conventional technologies such as multi-functional and synergistic applications.

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National Nuclear Security Administration